Anonymous Referee #3

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The authors present a multi-species time series of trace gas data from three flask stations in India. The data represent a very valuable contribution to this area of the world, which is currently poorly monitored, and the authors have analysed various aspects of the data (trends, gradients, etc) and covariance between species. However, the manuscript requires some revision to bolster some of the scientific conclusions that are made. If these comments can be addressed, the manuscript should be published.

General comments:

1. Introduction – There should be a comprehensive review of other measurement programs in South Asia – there is a description of CARIBIC and satellite-based studies, but there lacks a detailed description of other measurements (e.g., Bhattacharya et al., 2011, Ganesan et al, 2013 and Tiwari et al, 2014). At the moment it reads as though there are no other surface measurements (whether concurrently or previously) and while the authors discuss some very brief comparisons in the Results section, this needs to be brought forward into the Introduction. On page 7175 Line 9: 'Besides a lack of observation sites' should be written more accurately, which is that there are few observation sites in addition to those presented here, but this is not enough to constrain a large country like India.

[Response] Thanks a lot for your careful review and comments. Following your suggestions, we revised the second paragraph of the *Introduction* section and added a more detailed description of the surface atmospheric stations that have been recently established in India, including the stations in Sinhagad (18.35°N, 73.75°E, 1600m a.s.l.; Tiwari and Kumar, 2012; Tiwari et al., 2014), Mount Abu (24.60°N, 72.70°E, 1700m a.s.l.; S. Lal, personal communication), Ahmedabad (23.00°N, 72.50°E, 55m a.s.l.; Lal et al., 2015), Nainital (29.37°N, 79.45°E, 1958m a.s.l.; Kumar et al., 2010) and Darjeeling (27.03°N, 88.15°E, 2194m a.s.l.; Ganesan et al., 2013). Note that most of these stations started to measure GHG concentrations very recently (e.g. Sinhagad – since 2009; Ahmedabad – since 2013; Mount Abu – since 2013; Nainital – since 2006; Darjeeling – since 2011), and datasets are not always available. We also rephrased a few sentences in the second and third paragraphs accordingly (Lines 83–99, 101).

2. The authors compare their data to many other sites from NOAA and ICOS. While it is understandable that these measurements are directly linked to the authors and may overlap the time period of this study, there are measurements in India that should be compared to (see previous point), as these are very related to the conclusions made here (i.e. about regional sources, etc). Any comparisons made to other surface data are very minimal at present. The comparisons to CARIBIC, satellites, etc are important but to a lesser degree than other Indian surface observations.

[Response] Thanks a lot for your careful review and comments. In this study, we compared the flask measurements at HLE with KZM and WLG as well as those from the CONTRAIL and CARIBIC projects for several reasons: First, they all sample free-tropospheric air masses in northern mid-latitudes; Second, both HLE and the CARIBIC flights (and probably satellite measurements as well) show influences of the SW monsoon (and associated deep convection) on trace gas concentrations in the mid-to-high troposphere; Third, currently there is no ground station in India other than HLE that is representative of free tropospheric background concentrations over northern mid-latitudes. For N_2O and SF_6, we also compared gradients between PON, PBL and HLE to gradients between stations in Europe and the US, where GHG emissions are better known and relatively more accurate. Following your suggestions, we also referred to several previous papers on Indian surface observations (Bhattacharya et al., 2009; Ganesan et al., 2013; Tiwari et al., 2011, 2014) and added more discussions in *Section 3.1* (Lines 429–434, 494–497, 534–536).

3. The authors should be careful throughout the text to maintain that the mechanisms proposed for the various features in the data set are still speculative. This is a measurement-led study and without additional tools to quantitatively pinpoint the sources of air masses, these remain as hypotheses. An example of this would be on page 7187 line 21: "Moreover, the mean CH_4 seasonal cycle at HLE agrees well with the annual variation of convective precipitation over the Indian subcontinent (Fig. 5b), which is derived from ECMWF nudged Laboratoire de Météorologie Dynamique general circulation model (LMDz) (Hauglustaine et al., 2004). This agreement indicates that the summer maximum at HLE can be attributed to the enhanced biogenic CH_4 emissions from wetlands and rice paddies and deep convection that mixes surface emissions into the mid-to-upper troposphere." There is not enough information to say conclusively that biogenic emissions are responsible for the summer maximum without additional data (i.e. though models or isotopic data). So while the mechanism is proposed, it is stated too definitively. There are several statements like this throughout the text, which should be toned down and the authors should rephrase or remove statements such as this one.

[Response] Thanks a lot for your careful review and comments. In India, ruminant animals, natural wetlands and water-flooded rice paddies are the main sources of CH_4 emissions, accounting for ~40%, 15% and 16% of the total estimate. As we know, CH_4 emissions from ruminant animals do not show notable seasonality. By contrast, CH_4 emissions from natural wetlands and water-flooded rice paddies are greatly affected by climate conditions and subject to the seasonal variations of the Indian monsoon system. As illustrated in Fig. R1, emissions from wetlands and rice paddies show pronounced seasonality and have the maximum during July–September, exactly the same period when the SW monsoon prevails and the deep convection is most active. Therefore it is very likely that the summer maximum at HLE may be related to the enhanced biogenic CH_4 emissions from wetlands and rice paddies and deep convection that mixes surface emissions into the mid-to-upper troposphere. We agree that with the help of carbon isotopic measurements and/or chemical transport

model, we are able to further disentangle and quantify the contributions of meteorology and biogenic emissions to the CH_4 summer maximum at HLE. Following your suggestions, we added another panel to Fig. 5 and revised *Section 3.1.2* and *Conclusions* (Lines 472–483, 984) accordingly to clarify the statements.

4. Following up on the above statement, there are some sections, which are still quite speculative and not necessarily based on evidence and should be removed. These include: (a) Section 3.3 on elevated CH_4 and CO samples at PBL – There is not enough information to ascertain whether the samples at BKT are related to the samples at PBL. There would need to be a model simulation to show that the air mass at BKT on e.g., Sep 8 2009, arrived at PBL on Sep 16 2009. Otherwise it is too speculative and should be removed. (b) Discussion of bimodal H_2 on page 7196 line 17 – it is speculated the biomass burning from each hemisphere is the source of the double peaks. But there is no evidence to show that is the case.

[Response] Thanks a lot for your careful review and comments. (a) For *Section 3.3*, we agree that the mechanisms we proposed for the abnormal CH_4 and CO events and the possible linkage between PBL and BKT during the SW monsoon season are speculative, and need further verification with model experiments. Following your suggestion, we revised the manuscript and toned down the statements (Lines 944–952). (b) For discussion of the bimodal H_2 seasonal cycle at PBL, following your suggestion, we revised the manuscript and removed the sentences that are not accurate (Lines 774–776).

5. Many conclusions are drawn about Indian fluxes using HLE. However, from the text and looking at the trajectories, HLE mainly samples air from Africa and the Middle East. There are only a few trajectories that sample Indian air masses. It seems that the conclusions to the HLE data (with regards to Indian sources) should be changed to reflect this. Can HLE be used to discuss Indian sources?

[Response] Thanks a lot for your careful review and comments. In this study, we chose HLE as a reference station, and used the concentration gradients between PON, PBL and HLE to discuss the possible GHG sources in the Indian subcontinent. As we stated in *Section 2.1*, HLE (32.780 °N, 78.960 °E, 4517 m a.s.l.) is a high-altitude station situated in the western Himalayas. It dominantly samples mid-tropospheric air masses that pass over northern Africa and the Middle East throughout the year, and those coming from South and Southeast Asia during the SW monsoon season (also see the revised Fig. 1 colored by altitudes of back-trajectories). Therefore it is representative of free mid-troposphere background concentrations over northern mid-latitudes, rather than Indian air masses. That's why we chose this station as a background station, and used the concentration gradients between PON, PBL and HLE to infer whether or not there are substantial GHG emissions over South Asia (see details in *Section 3.1* for each species).

6. There appear to be some discrepancies in the text. The use of CARIBIC data and other remotely sensed data seems contradictory in places. In the discussion for SF_6, it states that the

CARIBIC samples are more representative of westerly jet transport rather than the SW monsoon. However, CARIBIC is used in the discussion for all other species in the context of Indian sources. It would also be useful to see trajectories for the comparison data to know whether they are sampling the same air masses.

[Response] Thanks a lot for your careful review and comments. In the discussion for SF_6, we compared the SF_6 seasonal cycle observed at HLE with that derived from CARIBIC flights. We cited Schuck et al. (2010), in which flask samples were taken during flights between Frankfurt and Chennai in 2008 over the domain 10–40°N, 50–80°E at flight altitudes 8–12.5km. As described in *Section 3* and *Section 5.3* in Schuck et al. (2010), the flask samples taken in summer were influenced by the monsoon anticyclone in the upper troposphere, as well as the westerly subtropical jet (see also Fig. R4). The summer maxima in CH_4 and N_2O by the CARIBIC flights were related to the monsoon anticyclone that can trap pollution uplifted by deep convection from the surface, and the back-trajectories analyses also show that samples taken over the monsoon region have ground contact (Fig. R4). The summer maximum in SF_6 was related to air samples collected north of 20°N along the flight routes, where air masses are more influenced by the westerly subtropical jet (and a smaller anticyclone located over the Arabian Peninsula embedded in it, see *Section 5.1* in Schuck et al. (2010) and Fig. 1 in Krishnamurti et al. (2008)) rather than the deep convection in the monsoon region.

As a high-altitude mountain station in the mid-troposphere (4517 m), HLE also samples polluted air masses uplifted by the deep convection in the monsoon region during summer as the CARIBIC flights do, but it is not influenced by the westerly subtropical jet located in the upper troposphere (also clearly seen by the colors of back-trajectories in Fig. R4). Therefore the summer enhancements of SF_6 observed by the CARIBIC flights are not detected by the flask measurements at HLE. Following your suggestion, we calculated and plotted back-trajectories for the CARIBIC flights investigated in Schuck et al. (2010) and added it to supplement (Fig. S8). We also revised the manuscript accordingly for clarification (Lines 630–636).

7. PON is located in a large urban area. While sampling is done between 1200-1800, the site would still be affected by local emissions. The analysis using PON for gradients between other sites could potentially be complicated by the fact that the site is impacted by local emissions. Therefore, PON may not be the best site to use for trend analysis. Can the authors comment on this? Could CO be used as a tracer for local emissions?

[Response] Thanks a lot for your careful review and comments. We agree that PON can be influenced by local emissions. Although the highway nearby has a low traffic flow, in-situ measurements at PON (not presented in this paper) do show that this site is heavily polluted by local emissions during nighttime. Therefore, we used two approaches to minimize the influences of local GHG sources/sinks. First, we took flask air samples at PON between 12:00 and 18:00 local time (actually 97% between 12:00 and 14:00 local time), when the sea

breeze moves towards land and the boundary layer air is well mixed (see *Section 2.1* for details). The recirculation of continental air mass during the sea breeze period should average regional influences, even though the footprint of PON is less than those of HLE and PBL. Second, when we performed the CCGVU curve-fitting, any data lying outside 3SD of the residuals were regarded as outliers and discarded from the time series, and this procedure was repeated until no outliers were identified (Harris et al., 2000; Zhang et al., 2007) (see *Section 2.3.1* for details). These outliers were likely a result of pollution by local emissions and not representative of regional background concentrations (denoted by crosses in each panel of Fig. 2, 4, 6, 8, 10 and 12). We believe that through the two approaches the local influences at PON should be sufficiently minimized.

Further, following your suggestion, we tried to use CO as a tracer for local emissions and filtered time series of other species by CO outliers. That means, for each species (other than CO), we removed the samples with abnormal CO values before the curve-fitting procedures. As shown in Table R1 and Fig. R2, filtering time series by CO outliers does not make significant difference to the trends, seasonal cycles and mean annual gradients (relative to HLE) for other species at this station. On the other hand, however, this filtering approach may substantially decrease the number of samples used to fit the smooth curve (e.g. ~38% for CH_4) and result in larger data gaps (Table R1, Fig. R2), probably compromising reliability of the analyses. Therefore finally we didn't use CO as a tracer of local emissions for additional filtering.

Specific comments:

Page 7173 line 15: change ‘dominant’ to ‘likely’ source of emissions

[Response] Following your suggestion, we revised it.

Page 7173 line 18-19: sentence needs restructuring. Suggest ‘to better constrain the GHG budget at regional and continental scales’

[Response] Following your suggestion, we revised it.

Introduction first paragraph: Text should state that the emissions from EDGAR, etc are using bottom-up methods, which generally have large uncertainties, and therefore top-down studies are needed as well.

[Response] Thanks for your suggestion. We agree that both bottom-up and top-down methods are important for estimation of GHG budgets. As we stated in the second paragraph of Introduction (please see the revised manuscript, Lines 65–75), current estimates of GHG budgets in India from both methods have larger uncertainties compared to Europe and North America.

Page 7174 line 11: what percent are natural emissions?

[Response] The natural CH_4 sources over land include emissions from wetlands, biomass burning and termites. Based on a combined dataset of (1) anthropogenic emissions from EDGARv4.2 FT2010 product, (2) wetland emissions from outputs of a global vegetation model (BIOME4-TG, Kaplan et al., 2006), (3) biomass burning emissions from Global Fire Emissions Database GFEDv3.0 product, and (4) termite emissions (Sanderson, 1996), we estimated that the natural emissions accounted for ~18% of the total CH_4 emissions over India.

The natural N_2O sources over land include emissions from uncultivated ecosystems, as well as biomass burning. Based on a combined dataset of (1) anthropogenic emissions from EDGARv4.2 FT2010 product, (2) fluxes from uncultivated ecosystems from the empirical of Bouwman et al. (2002), (3) biomass burning emissions from Global Fire Emissions Database GFEDv3.0 product, we estimated that the natural emissions accounted for ~53% of the total N_2O emissions over India. Note that both of them are rough estimates and subject to large uncertainties.

Page 7174 lines 15 – 17: Monitoring is not required by the UNFCCC.

[Response] Following your suggestion, we removed it.

Page 7175 lines 1-23: Following general comment above, a review of other surface measurements in India is needed in this paragraph. 'Besides a lack of observation sites' I agree that sites are sparse but they are not discussed.

[Response] Following your suggestions, we revised the second paragraph of the *Introduction* section and added a more detailed description of the surface atmospheric stations that have been recently established in India. We also rephrased a few sentences in the second and third paragraphs accordingly. Please see the revised manuscript Lines 83–101.

Page 7176 line 25: It is not possible to tell from the trajectories, what altitude these air masses originated from. HLE, for example, likely does not always sample surface emissions. It would be useful to see what altitude all of the sites are sampling. Also this would make comparison to aircraft observations easier to interpret.

[Response] Following your suggestion, we revised Fig. 1 and colored the back-trajectories by elevations of air masses instead of CO_2 levels. As we stated in *Section 2.1*, Fig. 1 shows that HLE dominantly samples mid-tropospheric air masses that pass over northern Africa and the Middle East throughout the year, and those coming from South and Southeast Asia during the SW monsoon season. Therefore it is representative of free mid-troposphere background concentrations over northern mid-latitudes.

Page 7177 line 17: manuscripts in preparation should not be cited

[Response] Following your suggestion, we removed it.

Page 7177 line 21: It looks like there are very few HLE trajectories coming from South Asia. Can the authors comment on the use of this site for regional work?

[Response] Thanks a lot for your careful review and comments. Please refer to our response to your general comment #5.

Page 7178 line 6: Do the sea breezes necessarily imply that they will be clean air masses? For example during the SW monsoon, the sea breeze will be a local effect on a dominant southwesterly flow. At PON, does this mean that air masses could still contain "local" emissions albeit the wind direction coming from the sea?

[Response] Thanks a lot for your careful review and comments. Please refer to our response to your general comment #7.

Page 7178 lines 5-7: Can CO be used as a tracer of local emissions for additional filtering for local emissions?

[Response] Thanks a lot for your careful review and comments. Please refer to our response to your general comment #7.

Page 7179 lines 19-23: Are flasks filled manually or automatically at a given time? Does an operator decide when conditions are correct for filling?

[Response] Flasks are flushed manually at a rate of 4–5 L min^{-1} for at least 10 minutes, corresponding to 40–50 L in total (i.e., flushing 40 times the volume of a flask). The operator decides how long flasks are flushed but the minimum required flushing time is 10 minutes.

Page 7180 line 19: Is there any impact of CO_2 on N_2O concentrations through this method? It is known that CO_2 can "dope" the signal for N_2O on an ECD.

[Response] Yes, the coelution of CO_2 is a concern in the gas chromatographic measurement of N_2O because CO_2 (with the same molecular weight as N_2O) reacts with intermediates of N_2O ionization in the ECD, thus enhancing the N_2O signal (Schmidt et al., 2001). We applied the procedures described in Lopez (2012) to solve the problem.

Page 7180: No description of the ECD or RDG setup (temps, flow rates) or information about carrier gases or calibration scales. Perhaps a table could provide all of the measurement info for each detector concisely.

[Response] Following your suggestion, we added in *Section 2.2.2* more details of the ECD and RGD setup as well as the carrier gases (Lines 259–272). We also added a table to list the configurations and parameters in the GC system (Table S1).

Page 7181 line 28: What are sampling uncertainties due to? local influence, human error?

[Response] The sampling uncertainties are mostly due to leakage of flask samplers or human errors (e.g., an operator who is not sufficiently trained yet and does not strictly follow the sampling or analysis protocol).

Page 7183 lines 2-3: Were the biases corrected?

[Response] No, the biases were not corrected as we don't know the true values.

Page 7184 line 12: 'additionally' should be 'additional'

[Response] Following your suggestion, we revised it.

Page 7185 line 12: HLE and CONTRAIL flights over New Delhi would likely be sampling different air masses, with HLE mostly seeming to sample air from the Middle East. Which altitude in the CONTRAIL profile represents the same air mass as HLE? Trajectories would be useful.

[Response] Following your suggestion, we computed and plotted five-day back-trajectories for all sampling hours of the in-situ CO_2 measurements over New Delhi by the CONTRAIL project (2006–2010). As shown in Fig. R3, the CONTRAIL flights at 3–6 km over New

Delhi sample the free-tropospheric air masses that pass over northern Africa and the Middle East throughout the year, and those coming from South Asia, Southeast Asia and the Arabian Sea during the SW monsoon season (Fig. R3a-c). They generally represents the same air mass as HLE (Fig. R3d), and do not show much difference across different altitude bands. We also added this figure in the Supplement (Fig. S7) and revised the main text accordingly (please see the revised manuscript Lines 395–396).

Page 7185 line 25: Again, it does not seem that HLE received many air masses from South Asia from the trajectories in Figure 1

[Response] Thanks a lot for your careful review and comment. Please refer to our response to your comment #5.

Page 7185 line 28: KZM and WLG, if they are more affected by northern air, then they would show a greater amplitude of the seasonal cycle. Can the authors comment?

[Response] Like HLE, KZM and WLG are high-altitude mountain stations, representative of the free-tropospheric background concentrations in northern mid-latitudes (Fig S4–5). As we know, the seasonal cycle of atmospheric CO_2 at surface observation stations in the Northern Hemisphere is driven primarily by net ecosystem production (NEP) fluxes from terrestrial ecosystem (Keeling et al., 1989; Manning, 1993; Erickson et al., 1996). In the far north, the amplitude of the CO_2 seasonal cycle ranges 15–20 ppm, and it diminishes southwards owing to the diminishing seasonality of plant activity towards the tropics (Keeling et al., 1996). As we mentioned in the manuscript, KZM and WLG are more influenced by northern air masses passing over Siberia with stronger seasonal CO_2 fluctuations, therefore CO_2 measurements at the two stations show larger amplitudes of seasonal cycles (as shown in Fig. 3b). The latitudinal gradients in the amplitude of the CO_2 seasonal cycle is also well illustrated in a 3D distribution of NOAA CO_2 Marine Boundary Layer (MBL) Reference (http://www.esrl.noaa.gov/gmd/ccgg/mbl/).

Page 7187 line 13: it seems that CARIBIC samples during the monsoon would not take one month to mix during this time of deep convection. Can the authors justify this statement? Also, why does vertical mixing lead to a larger seasonal cycle amplitude than HLE?

[Response] For the first question, we agree that during the SW monsoon, surface air masses with enhanced levels of trace gases are lifted by the strong deep convection over the Indian continent and rapidly mixed into the upper troposphere. Actually from Fig. 5a we can't tell a phase shift in CH_4 seasonal cycle by a lag of one month between the CARIBIC observations and HLE. For the CARIBC observations, the visible CH_4 seasonal maximum in September is not significant due to large errors of estimates in August and September (Fig. 5a). We removed this statement in the manuscript accordingly.

For the second question, during the SW monsoon period an anticyclone develops in the upper troposphere (Krishnamurti and Bhalme, 1976). Observations have shown persistent maxima

of many trace gases in the monsoon anticyclone during summer (Park et al., 2004; 2007), probably due to vertical transport of surface air masses by deep convection and subsequent accumulation and confinement of pollutants within the strong, closed circulation of the anticyclone (Li et al., 2005; Randel and Park, 2006). Randel and Park (2006) also showed that the monsoon anticyclone can trap air masses for up to several weeks depending on altitude, with more effective confinement occurring at higher altitudes. Since the CARIBIC flights sampled at altitudes 8–12.5km over this region (Schuck et al., 2010), we observe a larger amplitude of the CH_4 seasonal cycle than HLE.

Page 7187 line 16: Remove 'apparently'

[Response] Following your suggestion, we removed it.

Page 7187 lines 22-28: This discussion is too speculative and should be removed without further evidence (e.g., isotopic). There is not enough information to state that biogenic CH_4 emissions are responsible for the summer max at HLE. Furthermore, a model is needed to disentangle the meteorology from emissions.

[Response] Thanks a lot for your careful review and comments. Please refer to our response to your general comment #3.

Page 7187 line 29: be more specific - concentrations of trace gases would be enhanced at higher altitudes rather than the surface.

[Response] Following your suggestion, we revised it.

Page 7188 line 3: Earlier it is stated that KZM and WLG sample wetland emissions from the north. But here it is stated that their CH_4 increases are smaller because they are not influenced by deep convection. Does that necessarily imply that the increases will be smaller? There could be a large summer methane signal from wetlands.

[Response] The ground station measurements of CH_4 in the Northern Hemisphere usually show a summer minimum, predominantly due to oxidation of CH_4 by the OH radicals (Dlugokencky et al., 1994). The summer maxima observed at HLE, KZM and WLG likely result from transport of the air masses that are enriched in CH_4 and not yet consumed by OH before reaching the station. For KZM and WLG, the CH_4-enriched air masses are probably transported from Siberia with large wetland emissions in summer, and/or regional sources closer to the stations (Fang et al., 2013; also see back-trajectories in Fig. S4). Without deep convection during summer, at least the vertical transport of polluted air masses would be less efficient at the two stations, which could be one reason responsible for the smaller CH_4 enhancements compared to HLE. Indeed, further analyses (e.g. chemical transport model) are needed to resolve contribution of different sources and transport to the CH_4 enhancements at the three stations in summer. We revised the manuscript accordingly to make it clearer and precise (Lines 483–485).

Page 7188 line 10: Why does PON not sample surface emissions? The trajectories during July look like they pass over southern India.

[Response] Thanks a lot for your careful review and comments. We agree that this statement is not consistent with the back-trajectories at PON and removed it from the manuscript accordingly.

Page 7189 line 18: Why would it be argued that N_2O is 'more noisy than CO_2 and CH_4 due to regional sources synoptic variability'? Also, N_2O measurement has lower signal to noise (i.e. precision is lower than CO_2 and CH_4).

[Response] Thanks a lot for your careful review and comments. As you mentioned, the N_2O measurement has a lower signal-to-noise ratio. When we argue that the seasonal cycle of N_2O is noisier compared to CO_2 and CH_4 in the manuscript, it means the N_2O seasonal cycle has a larger uncertainty (i.e. lower precision, also indicated by the wide shaded area in Fig. 7). Given that the N_2O seasonal cycle is very small, synoptic events are more likely to mask the seasonal signal. As shown in Table 1, if we take the ratio of the seasonal cycle amplitude to the residual standard deviation (RSD, an indicator of synoptic variability) as a surrogate of the signal-to-noise ratio, we find that this ratio is significantly lower for N_2O (2.0, 1.5 and 2.0 for HLE, PON and PBL) than CO_2 (11.1, 1.9 and 7.1 for HLE, PON and PBL) and CH_4 (3.2, 3.6, 6.3 for HLE, PON and PBL). Following your suggestion, we revised the statement in the manuscript for clarification (Lines 542–543).

Page 7190 line 7: CARIBIC enrichment only during monsoon – why April-December 2008?

[Response] Following your suggestion, we revised the sentence to "Like CH_4, the N_2O enhancement at HLE during the summer monsoon period (June-September) is consistent with the aircraft flask measurements at flight altitudes 8–12.5 km from the CARIBIC project in 2008 (Schuck et al., 2010)" (Lines 558–561).

Page 7191 line 24: Even if there were no SF_6 emissions (rather than weak SF_6 emissions), this would imply that sites should follow the background. This still doesn't explain why there is a negative gradient.

[Response] The negative gradient between PON and HLE is likely due to the fact that HLE dominantly samples air masses passing over North Africa and the Middle East, where SF_6 emissions are substantial (Fig. R5). By contrast, PON receives air masses from the South India (during the boreal summer) and the northeast parts of the Indian subcontinent (during the boreal winter), which are much less polluted by SF_6. As PBL samples air masses from Southeast Asia and Southwest China (during the boreal winter) with notable SF_6 emissions (Fig. R5), the gradient between PBL and HLE is statistically insignificant. We revised the manuscript accordingly for clarification (Lines 601–603).

Page 7192 lines 14-18: It is mentioned here that CARIBIC samples different air masses to HLE. It is unclear therefore why the CARIBIC comparison is made for CH_4 and N_2O. This seems like a contradiction and so perhaps CARIBIC comparison should be removed for CH_4 and N_2O as well for HLE.

[Response] Thanks a lot for your careful review and comment. Please refer to our response to your general comment #6.

Page 7193 line 23: It is difficult to see a one month lag in Fig 11.

[Response] Please also refer to Table 1. The lag in the CO seasonal minimum between WLG and HLE is about 30 days. We added this information to the manuscript (Line 683).

Page 7193 line 16: Could the larger variability also be due to local sources?

[Response] Thanks a lot for your careful review and comment. Please refer to our response to your general comment #7.

Page 7196 lines 19-25: This discussion about bimodal H_2 seasonal cycle being due to biomass burning is very speculative and should be removed. There is not enough information or model runs to demonstrate that this is the case.

[Response] Following your suggestion, we revised the manuscript and removed the sentences that are not accurate (Lines 774–776).

Page 7197 line 21: Describe why anthropogenic CO emissions are lower in summer than winter?

[Response] The anthropogenic CO emissions in India are mainly contributed by residential energy use (57%) and agricultural waste burning (19%) (EDGAR v4.2). The anthropogenic CO emissions are lower in summer than in winter, mainly due to the less residential fuel use for heating (Streets et al., 2003). We added this information to the manuscript accordingly (Line 801).

Page 7198 line 3: Why would uplift contribute to maximum CH_4/CO ratio, as both species are uplifted together?

[Response] As we know, HLE is a high-altitude mountain station (4517m a.s.l.) and the CARIBIC measurements used for comparison in this study were taken at flight altitudes 8 – 12.5km (Schuck et al., 2010). Without the convective uplift that mixes the surface polluted air masses to the mid-to-upper troposphere during the SW monsoon, the summer maximum $\Delta CH_4/\Delta CO$ ratio would not be observed by HLE or the CARIBIC flights.

Section 3.3: This section is too speculative, as the appropriate model simulations have not been performed to assess whether the elevated events are related to elevations at BKT. With

the model simulation, linking the time and position of the elevated event at BKT with the time and position of the elevated event at PBL, this section should be removed.

[Response] We agree that the mechanism we proposed for the abnormal CH_4 and CO events and the possible linkage between PBL and BKT during the SW monsoon season are speculative, and need further verification with model experiments. Following your suggestion, we revised the manuscript and toned down the statements (Lines 944–952).

Conclusions page 7204 line 8 – The summertime maximum being attributed definitively to biogenic emissions is too speculative without other information. The authors should tone down the statement.

[Response] Following your suggestion, we revised the manuscript and tone down the statement accordingly (Lines 983–987, 992–996). Please refer to the response to general comment #3 for a more detailed discussion of the statement.

Figures 3, 5, 7 etc should show uncertainties, from measurement uncertainty, sampling uncertainty and if averaged into seasonal cycle, the variability in the seasonal cycle. This would provide an indication for the significance of the seasonal cycle. Some panels in figures contain uncertainties, some do not.

[Response] All the plots of seasonal cycles show uncertainties, including those in the supplementary document. For a few stations (e.g., HLE), the seasonal cycle of a species may be too small (relative to the scale of the plot) too be visible. We tried to improve the quality of these figures for better display.

Supplement - Back trajectories for comparison data (KZM, WLG) should also include trajectories for CARIBIC, etc.

[Response] Following your suggestion, we calculated and plotted back-trajectories for the CONTRAIL and CARIBIC measurements we used for comparison in this study (Fig. R3-4, also see Fig. S7–8 in the supplement).

Supplement - A description of KZM and WLG is needed. Are they mountain sites, etc?

[Response] Yes, they are high-altitude mountain stations. The geographic locations (latitude, longitude and altitude) of the two stations were given in the manuscript in *Section 3.1.1* (Lines 368–370).

Short title - Change to ’Five years (plural) of flask measurements’

[Response] We will revise it when resubmitting the paper.

References

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Tables

Table R1 Features of the smoothed fitting curves for flask measurements at PON (2007–2011). For each species, the smoothed curves are fitted to the data not filtered by CO outliers and the data filtered by CO outliers. The annual mean values and average peak-to-peak amplitude are calculated from the smoothed curve and mean season cycle, respectively. Uncertainty of each estimate is calculated from 1 s.d. of 1000 bootstrap replicates.

	Not CO filtered	CO filtered
CO_2 (ppm)		
N_{fit}	121	105
Annual mean 2007	386.6±0.9	386.5±1.1
Annual mean 2008	388.1±0.9	388.0±0.9
Annual mean 2009	389.0±0.6	388.4±0.8
Annual mean 2010	391.3±1.5	391.2±1.5
Annual gradient relative to HLE	2.9±1.2	2.6±1.4
Trend	1.7±0.1	1.7±0.1
RSD	4.0	4.1
Amplitude	7.6±1.4	7.8±1.6
D_{max}	111.0±13.4	116.0±14.1
D_{min}	327.0±54.3	327.0±55.8
CH_4 (ppb)		
N_{fit}	164	101
Annual mean 2007	1859.2±6.7	1854.2±5.9
Annual mean 2008	1856.1±10.4	1857.3±6.8
Annual mean 2009	1865.7±5.1	1855.5±6.2
Annual mean 2010	1876.9±9.1	1877.3±7.3
Annual gradient relative to HLE	37.4±10.7	34.0±11.0
Trend	9.4±0.1	9.0±0.1
RSD	34.4	19.8
Amplitude	124.1±10.2	127.8±11.4
D_{max}	337.0±6.1	331.0±5.4
D_{min}	189.0±10.7	192.0±9.8
N_2O (ppb)		
N_{fit}	137	110
Annual mean 2007	324.8±0.3	324.9±0.4
Annual mean 2008	326.3±0.3	326.3±0.3
Annual mean 2009	326.7±0.3	326.4±0.3
Annual mean 2010	327.1±0.5	327.0±0.5
Annual gradient relative to HLE	3.1±0.3	3.0±0.3
Trend	0.8±0.1	0.7±0.1
RSD	1.4	1.4
Amplitude	1.2±0.5	1.1±0.5

D_{max}	262.0±83.2	262.0±46.1
D_{min}	141.0±48.2	97.0±65.8
SF_6 (ppt)		
N_{fit}	174	139
Annual mean 2007	6.19±0.01	6.19±0.02
Annual mean 2008	6.49±0.02	6.49±0.02
Annual mean 2009	6.77±0.01	6.77±0.02
Annual mean 2010	7.08±0.02	7.08±0.02
Annual gradient relative to HLE	-0.06±0.03	-0.06±0.03
Trend	0.31±0.05	0.31±0.06
RSD	0.05	0.05
Amplitude	0.24±0.02	0.24±0.03
D_{max}	327.0±12.1	327.0±21.7
D_{min}	204.0±3.3	205.0±3.4
CO (ppb)		
N_{fit}	139	139
Annual mean 2007	200.5±7.8	200.5±7.8
Annual mean 2008	175.3±13.1	175.3±13.1
Annual mean 2009	174.3±4.8	174.3±4.8
Annual mean 2010	185.1±8.7	185.1±8.7
Annual gradient relative to HLE	82.4±10.7	82.4±10.7
Trend	0.4±0.1	0.4±0.1
RSD	32.0	32.0
Amplitude	78.2±11.6	78.2±11.6
D_{max}	4.0±160.2	4.0±160.2
D_{min}	238.0±46.1	238.0±46.1
H_2 (ppb)		
N_{fit}	140	120
Annual mean 2007	574.5±2.4	573.7±3.2
Annual mean 2008	558.2±5.3	558.3±5.1
Annual mean 2009	562.4±1.6	561.9±1.6
Annual mean 2010	563.9±2.3	563.0±2.5
Annual gradient relative to HLE	29.8±4.1	29.3±3.7
Trend	-1.3±0.1	-1.3±0.1
RSD	8.4	8.3
Amplitude	21.6±3.4	21.1±3.8
D_{max}	96.0±9.6	97.0±9.8
D_{min}	219.0±10.3	215.0±11.9

Figures

Figure R1 The mean CH_4 seasonal cycles observed at HLE and seasonal variations of CH_4 emissions from rice paddies and wetlands over the Indian subcontinent. The CH_4 emissions from rice paddies are extracted from a global emission map for the year 2010 (EDGAR v4.2), imposed by the seasonal variation on the basis of Matthews et al. (1991). The CH_4 emissions from wetlands are extracted from outputs of a global vegetation model (BIOME4-TG, Kaplan et al., 2006). The seasonal variation of deep convection over the Indian subcontinent is also presented, indicated by convective precipitation obtained from an LMDz simulation nudged with ECMWF reanalysis. The CH_4 emissions and convective precipitation are averaged over the domain of 10–35 °N, 70°–90°E to give a regional mean estimate.

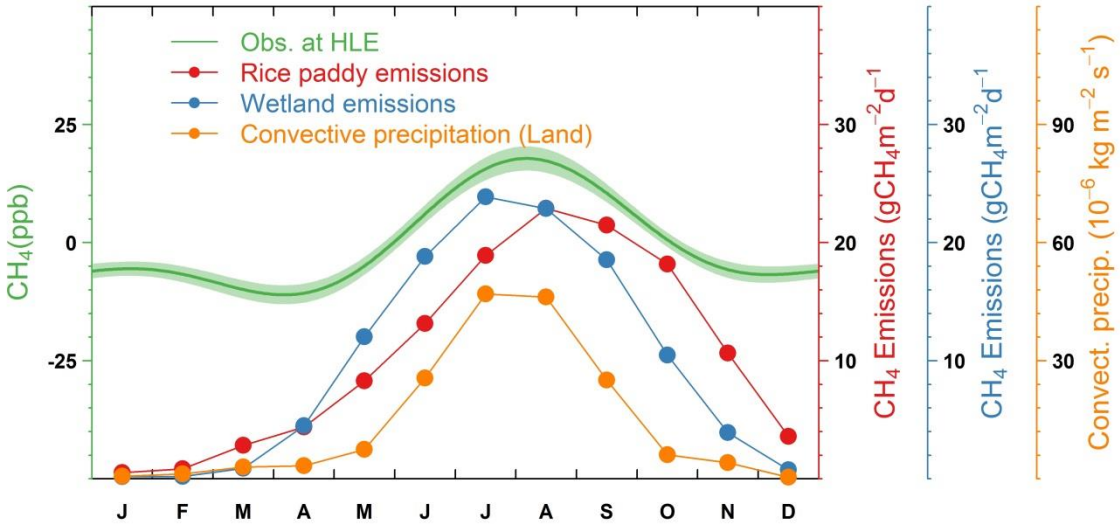
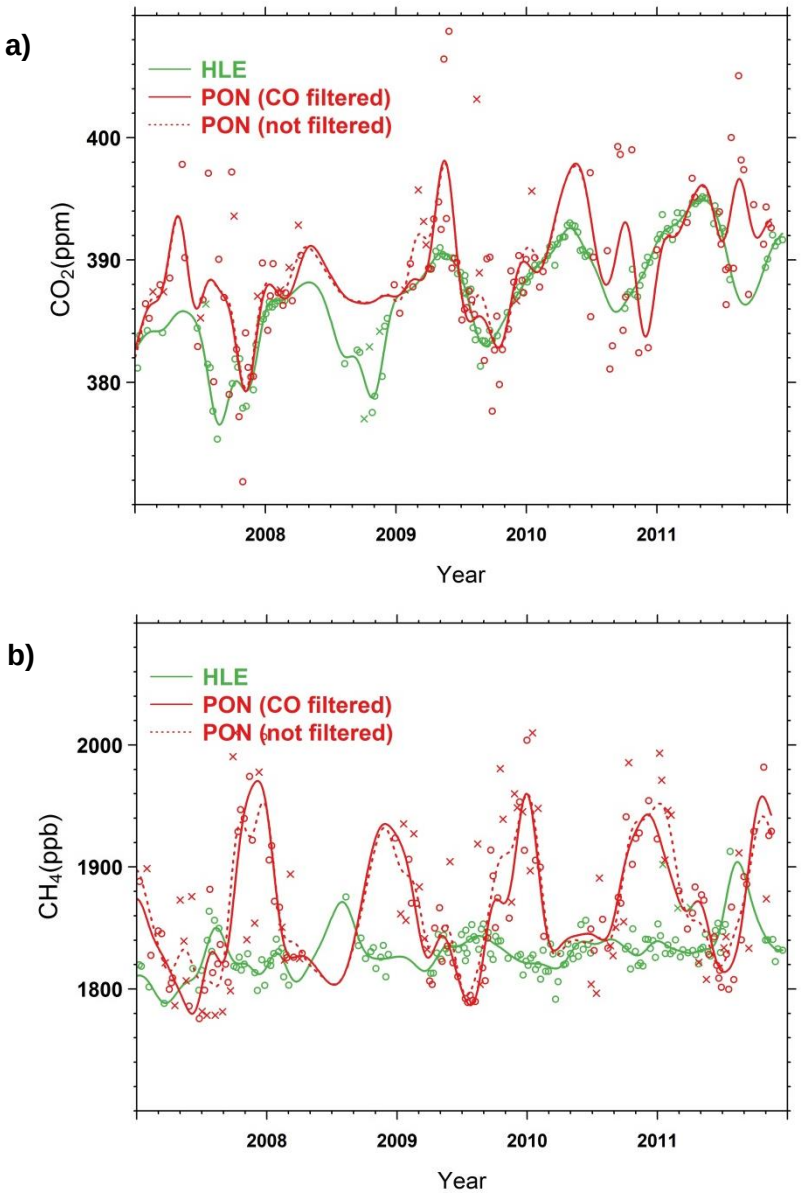
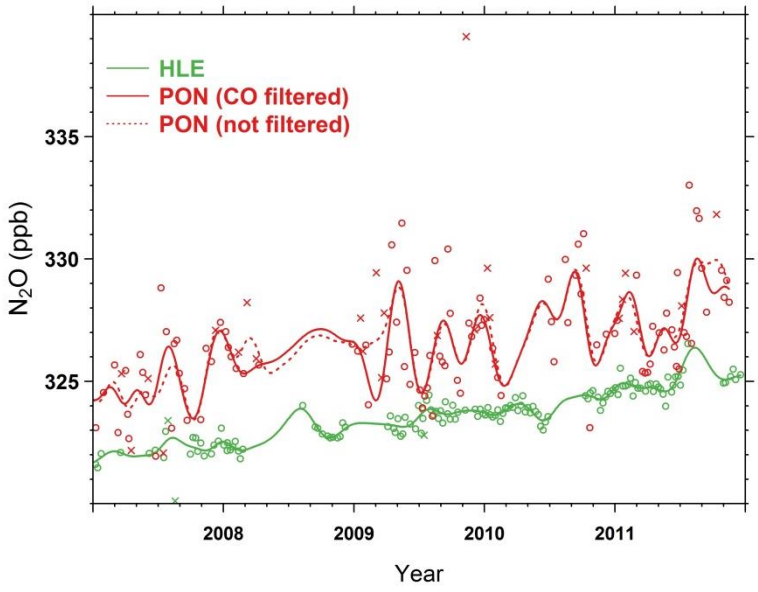


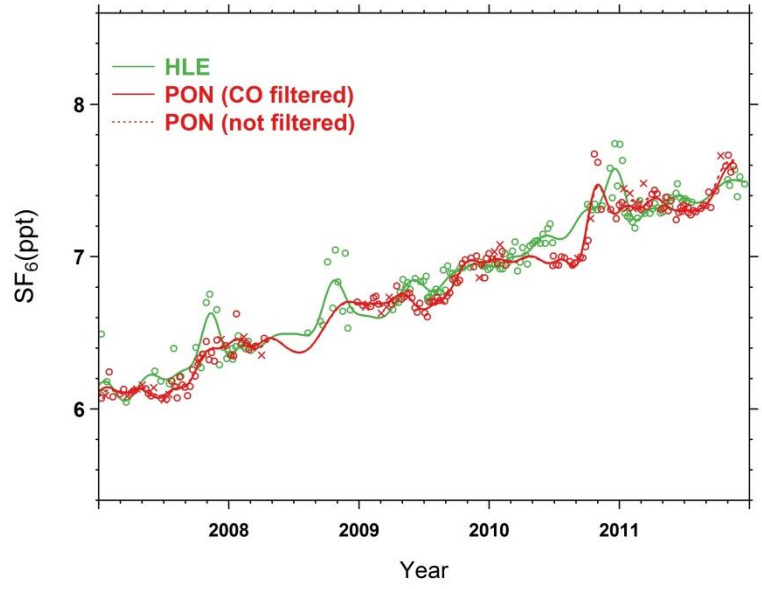
Figure R2 Time series of flask measurements at PON (2007–2011) with smoothed fitting curves for **(a)** CO_2, **(b)** CH_4, **(c)** N_2O, **(d)** SF_6 and **(e)** H_2. The open circles denote flask data used to fit the solid smoothed curves, while the crosses denote discarded flask data lying outside 3 times the residual standard deviations from the smoothed curve fits as well as those filtered by CO outliers. For PON, the solid (dotted) red line indicates the smoothed curve fitted to the data (not) filtered by CO outliers. The flask measurements at HLE and the corresponding smoothed fitting curve are also presented for comparison.



c)


HLE
PON (CO filtered)
PON (not filtered)
335
330
325
N2O (ppb)
2008
2009
2010
2011
Year

d)


HLE
PON (CO filtered)
PON (not filtered)
8
7
6
SF6(ppt)
2008
2009
2010
2011
Year

e)

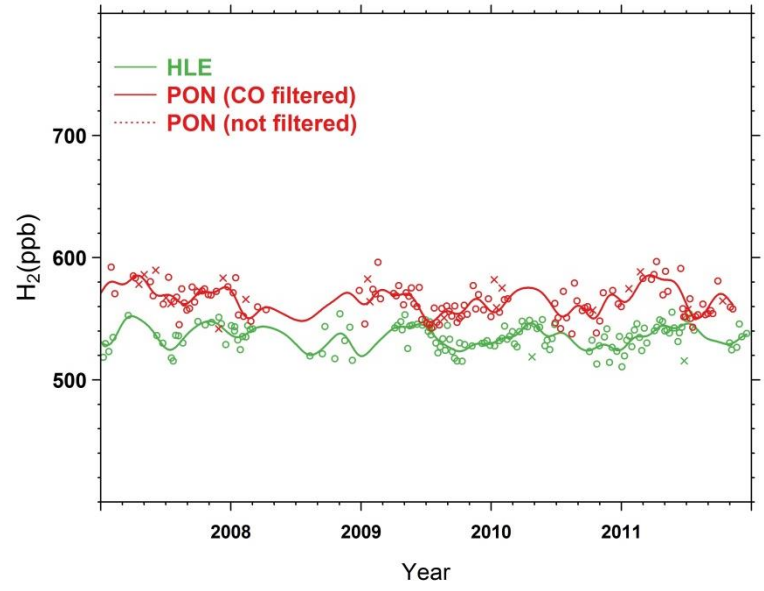
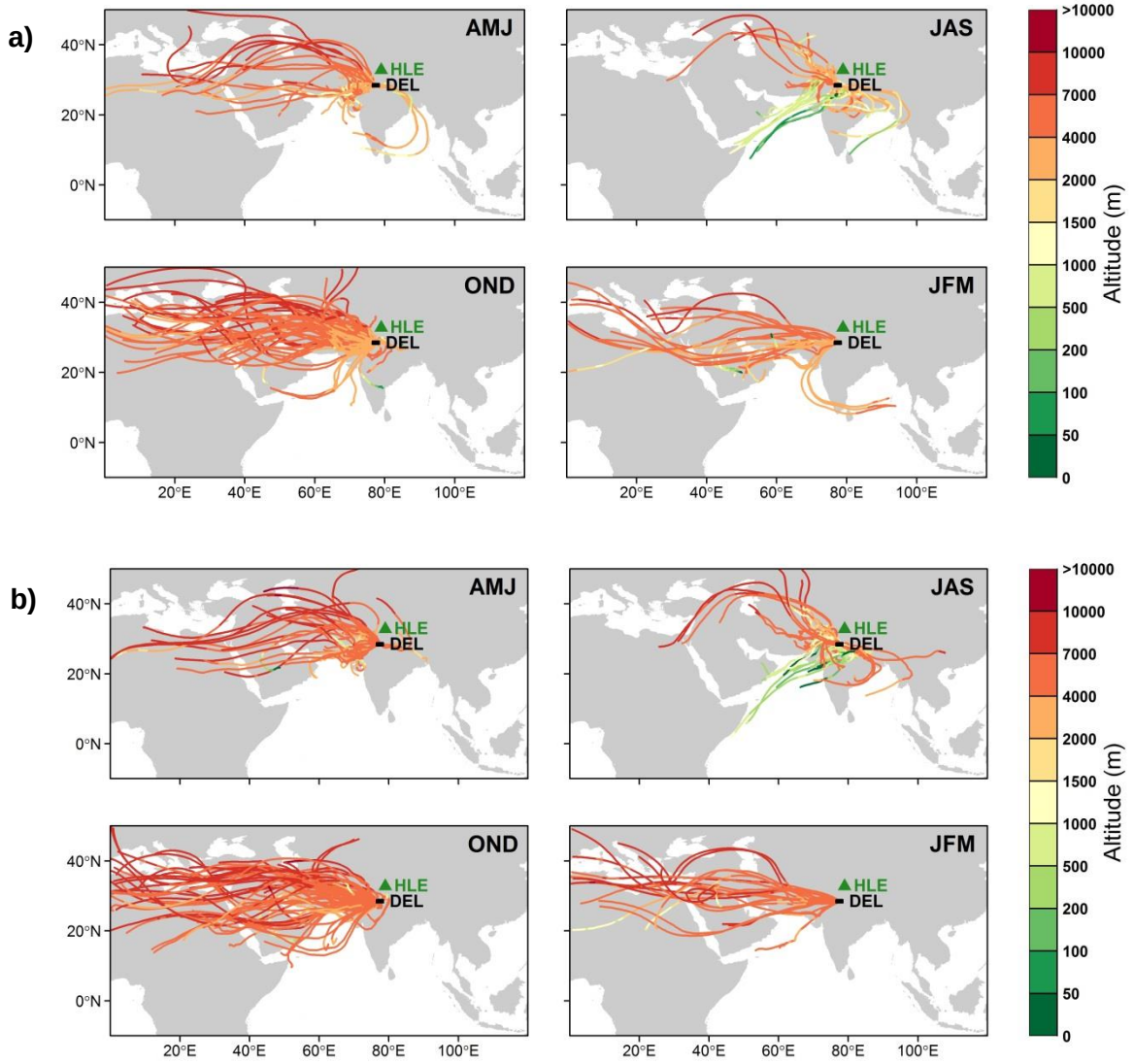
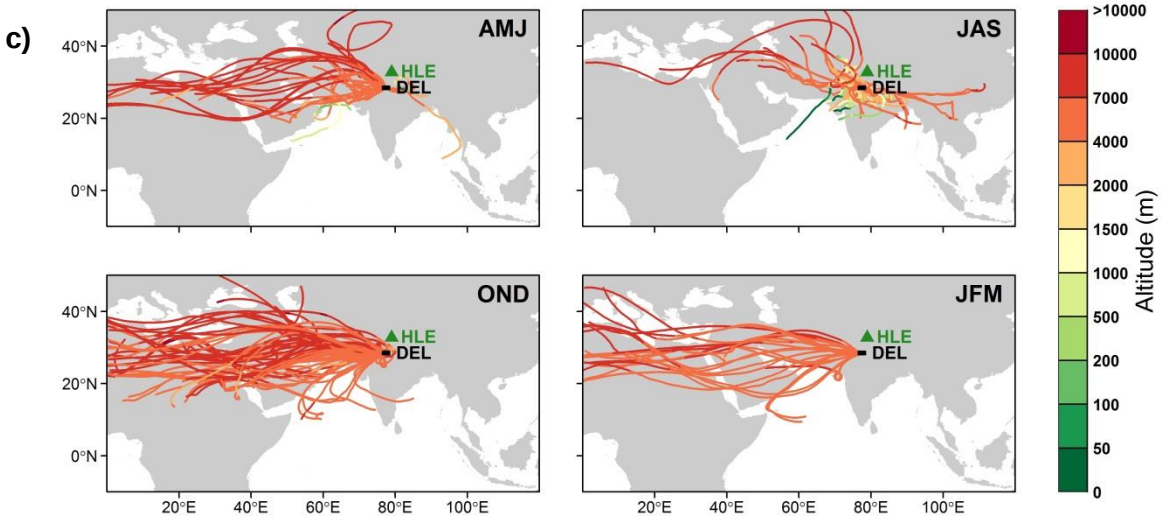

HLE
PON (CO filtered)
PON (not filtered)
700
600
500
H2(ppb)
2008
2009
2010
2011
Year

Figure R3 Five-day back-trajectories calculated for all sampling hours of the in-situ CO_2 measurements over New Delhi by the CONTRAIL project (2006–2010). Back-trajectories are computed and plotted at different altitude bands: **(a)** 3–4 km, **(b)** 4–5 km, and **(c)** 5–6 km. For comparison, back-trajectories for all sampling dates of the flask measurements at HLE (2007–2011) are also presented in **(d)**. All back-trajectories are colored by the elevation of air masses at hourly time step.




c)
AMJ
JAS
OND
JFM
HLE
DEL
40°N
20°N
0°N
20°E
40°E
60°E
80°E
100°E
>10000
10000
7000
4000
2000
1500
1000
500
200
100
50
0
Altitude (m)

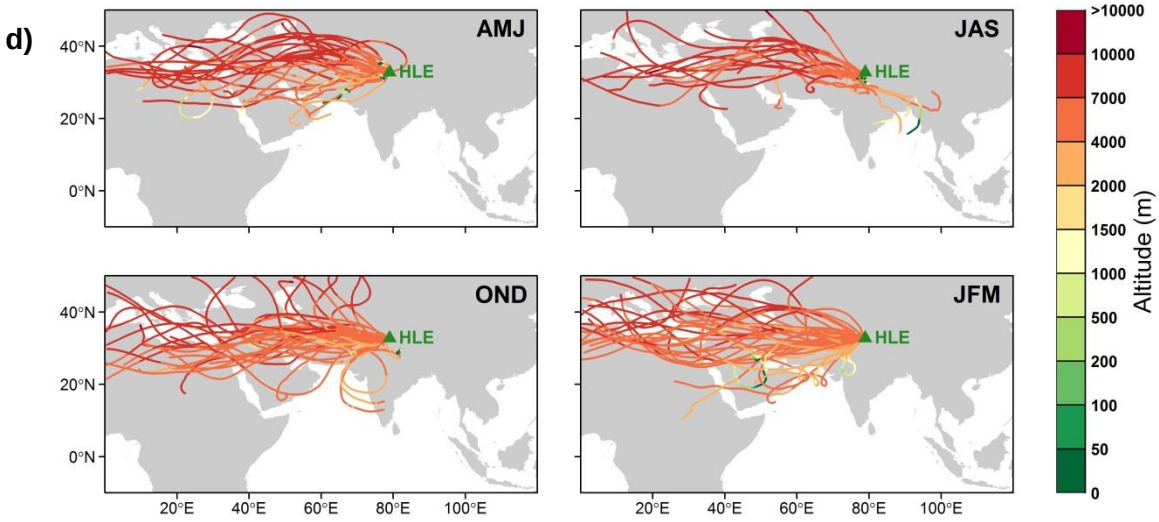

d)
AMJ
JAS
OND
JFM
HLE
40°N
20°N
0°N
20°E
40°E
60°E
80°E
100°E
>10000
10000
7000
4000
2000
1500
1000
500
200
100
50
0
Altitude (m)

Figure R4 (a) Five-day back-trajectories calculated for all sampling hours of the flask measurements by the CARIBIC flight between Frankfurt and Chennai at flight altitudes 8–12.5 km for the year 2008. The box indicates the domain of 10–40°N, 50–80°E, where flask samples within it were investigated in Schuck et al. (2010). **(b)** Five-day back-trajectories calculated for all sampling dates over the period 2007–2011 at HLE. All back-trajectories are colored by the elevation of air masses at hourly time step.

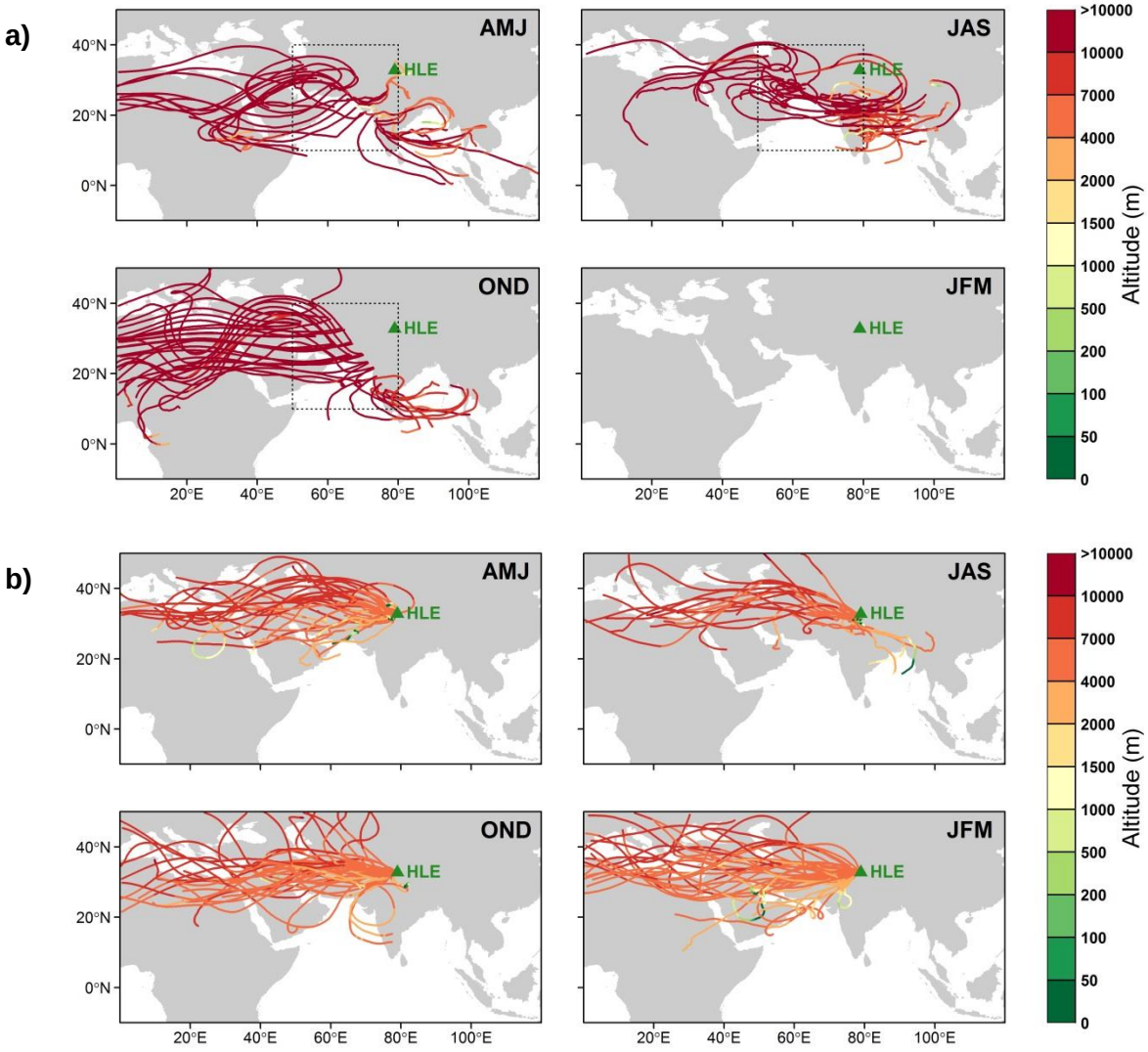


Figure R5 The map of SF_6 emissions for the year 2010 based on the EDGARv4.2 FT2010 dataset. This map is produced by EDGAR and can be downloaded from the website (http://edgar.jrc.ec.europa.eu/).